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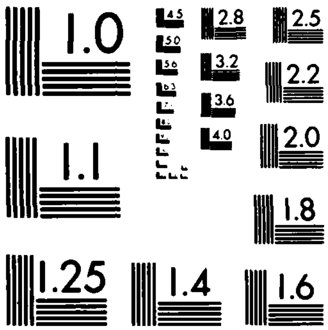
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DIRECT SYNTHESIS AND OPTIMIZATION OF RARE EARTH TRANSITION METAL PERMANENT MAGNET SYSTEMS

Final Technical Report

Professor F. J. Cadieu

15 September 1986

U. S. Army Research Office
Contract Number
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Queens College of CUNY
Flushing, NY 11367
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1515 Broadway
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AD-A172 955

REPORT DOCUMENTATION PAGE		READ INSTRUCTIONS BEFORE COMPLETING FORM
1. REPORT NUMBER: ARO 19743.9-MS	2. GOVT ACCESSION NO.: N/A	3. RECIPIENT'S CATALOG NUMBER: N/A
4. TITLE (and Subtitle): DIRECT SYNTHESIS AND OPTIMIZATION OF RARE EARTH TRANSITION METAL PERMANENT MAGNET SYSTEMS Final Technical Report		5. TYPE OF REPORT & PERIOD COVERED: Final Technical Report 1 August 1983-31 July 1986
		6. PERFORMING ORG. REPORT NUMBER
7. AUTHOR(s): F. J. Cadieu		8. CONTRACT OR GRANT NUMBER(s): DAAG29-83-K-0114
9. PERFORMING ORGANIZATION NAME AND ADDRESS: Department of Physics Queens College of CUNY Flushing, NY 11367 & Research Foundation of CUNY, 1515 Broadway New York, NY 10036		10. PROGRAM ELEMENT, PROJECT, TASK AREA & WORK UNIT NUMBERS: N/A
11. CONTROLLING OFFICE NAME AND ADDRESS: U. S. Army Research Office Post Office Box 12211 Research Triangle Park, NC 27709		12. REPORT DATE: 15 September 1986
		13. NUMBER OF PAGES: 42
14. MONITORING AGENCY NAME & ADDRESS (if different from Controlling Office)		15. SECURITY CLASS. (of this report): Unclassified
		15a. DECLASSIFICATION/DOWNGRADING SCHEDULE

16. DISTRIBUTION STATEMENT (of this Report)

Approved for public release; distribution unlimited.

17. DISTRIBUTION STATEMENT (of the abstract entered in Block 20, if different from Report)

NA

18. SUPPLEMENTARY NOTES

The view, opinions, and/or findings contained in this report are those of the author(s) and should not be construed as an official Department of the Army position, policy, or decision, unless so designated by other documentation.

19. KEY WORDS (Continue on reverse side if necessary and identify by block number)

Rare Earth Permanent Magnet Systems,
Samarium Cobalt
Neodymium Iron Boron
Samarium Titanium Iron
Permanent Magnet Films
Sputtered Magnetic Films
Sputtered Films

20. ABSTRACT (Continue on reverse side if necessary and identify by block number)

Special sputtering methods have been used to synthesize rare earth transition metal films such as $SmCo_5$, $Nd_2Fe_{14}B$, and several new compounds in the Sm-Ti-Fe system. Films have been synthesized with different crystal textures by varying the sputtering conditions. The magnetic properties observed have been found to be strongly dependent on the film texture. For example, $SmCo_5$ films with a (200) texture exhibit intrinsic

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20. ABSTRACT (continued)

Coercive force

coercive forces, $_{i}H_{c}$, of 8 kOe, while (110) textured films show $_{i}H_{c}$ values of about 18 kOe. For the $Nd_2Fe_{14}B$ compound films have been made which exhibit an easy axis of magnetization either in the film plane or out of the film plane depending upon the crystal texturing of the films. In the film plane static energy products up to 21 MG-Oe have been observed in as sputtered films without any subsequent heat treatments in certain rare earth transition metal systems. For films containing only Sm, Ti, and Fe room temperature $_{i}H_{c}$ values up to 24 kOe have been observed. Films have also been made such that the magnetic properties are highly anisotropic in the plane of the film for certain amorphous systems. In the film plane anisotropy constants up to 2.6 x 10^6 ergs/cm^3 have been observed for magnetizing films in two orthogonal directions within the film plane. In two Fe based systems, large perpendicular anisotropy has been observed. For a $Sm(Ti,Fe)_2$ compound effective perpendicular anisotropy constants up to 4 x 10^6 erg/cm^3 have been observed. For $Nd_2Fe_{14}B$ high values of $_{i}H_{c}$ up to 14 kOe have been observed for magnetometer fields up to 20 kOe applied perpendicular to the film planes. For magnetometer fields applied perpendicular to the film plane, the as measured magnetization curves continue flat well into the 2nd quadrant. The hysteresis loops when corrected for demagnetization effects are highly rectangular in this case. High field data will be needed to fully characterize these films since only minor hysteresis loops have been observed for applied fields of 20 kOe. For $Nd_2Fe_{14}B$ films effective perpendicular anisotropy constants up to 1.2 x 10^7 erg/cm^3 have been observed.

Coercive force

The incorporation of the $_{i}H_{c}$ magnetic films that have been sputter synthesized into several types of micron scale device geometries has been considered. Currently a patent application titled "Method of Making a Miniature Scale Periodic Permanent Magnet Array and Miniature Scaled Periodic Permanent Magnet Array So Formed". The objective and result of this patent is the fabrication of micron scale (1 micron = 10 meter) periodic permanent magnet arrays which can be magnetized by external magnetic fields which have a large spatial extent when compared to the dimensions of individual elements of the array. Certain cladded magnet geometries which make essential use of magnetic films which exhibit in the film plane anisotropy and films which exhibit perpendicular anisotropy as elements of the same device have also been considered.

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DIRECT SYNTHESIS AND OPTIMIZATION OF RARE EARTH TRANSITION METAL PERMANENT MAGNET SYSTEMS
Final Technical Report

F. J. Cadieu
Department of Physics
Queens College of the City University of New York

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DIRECT SYNTHESIS AND OPTIMIZATION OF RARE EARTH TRANSITION METAL PERMANENT MAGNET SYSTEMS: FINAL TECHNICAL REPORT

Professor F. J. Cadieu

Abstract

Special sputtering methods have been used to synthesize rare earth transition metal films such as $SmCo_5$, $Nd_2Fe_{14}B$, and several new compounds in the Sm-Ti-Fe system. Films have been synthesized with different crystal textures by varying the sputtering conditions. The magnetic properties observed have been found to be strongly dependent on the film texture. For example, $SmCo_5$ films with a (200) texture exhibit intrinsic coercive forces, ${}_iH_c$, of 8 kOe, while (110) textured films show ${}_iH_c$ values of about 18 kOe. For the $Nd_2Fe_{14}B$ compound films have been made which exhibit an easy axis of magnetization either in the film plane or out of the film plane depending upon the crystal texturing of the films. In the film plane static energy products up to 21 MG-Oe have been observed in as sputtered films without any subsequent heat treatments in certain rare earth transition metal systems. For films containing only Sm, Ti, and Fe room temperature ${}_iH_c$ values up to 24 kOe have been observed. Films have also been made such that the magnetic properties are highly anisotropic in the plane of the film for certain amorphous systems. In the film plane anisotropy constants up to 2.6×10^6 ergs/cm^3 have been observed for magnetizing films in two orthogonal directions within the film plane. In two Fe based systems, large perpendicular anisotropy has been observed. For a $Sm(Ti,Fe)_2$ compound effective perpendicular anisotropy constants up to 4×10^6 erg/cm^3 have been observed. For $Nd_2Fe_{14}B$ high values of ${}_iH_c$ up to 14 kOe have been observed for magnetometer fields up to 20 kOe applied perpendicular to the film planes. For magnetometer fields applied perpendicular to the film plane, the as measured magnetization curves continue flat well into the 2nd quadrant. The hysteresis loops when corrected for demagnetization effects are highly rectangular in this case. High field data will be needed to fully characterize these films since only minor hysteresis loops have been observed for applied fields of 20 kOe. For $Nd_2Fe_{14}B$ films effective perpendicular anisotropy constants up to 1.2×10^7 erg/cm^3 have been observed.

The incorporation of the ${}_iH_c$ magnetic films that have been sputter synthesized into several types of micron scale device geometries has been considered. Currently a patent application titled "Method of Making a Miniature Scale Periodic Permanent Magnet Array and Miniature Scaled Periodic Permanent Magnet Array So Formed". The objective and result of this patent is the fabrication of micron scale (1 micron = 10 meter) periodic permanent magnet arrays which can be magnetized by external magnetic fields which have a large spatial extent when compared to the dimensions of individual elements of the array. Certain cladded magnet geometries which make essential use of magnetic films which exhibit in the film plane anisotropy and films which exhibit perpendicular anisotropy as elements of the same device have also been considered.

INTRODUCTION

The magnetic properties of the systems synthesized fall into distinctly different classifications ranging from high intrinsic coercive force systems, high in the film plane energy product systems, films which exhibit perpendicular anisotropy, and films which exhibit a large degree of anisotropy within the film plane. In many cases the magnetic properties of the same system can be made to change dramatically by a variation of the sputtering conditions. For certain systems we have been able to synthesize the same crystal phase with different crystal texturing with consequently different magnetic properties. Some recent results for each of the following areas will be discussed: (1) High Energy Product Co Based Films, (2) High ${}_iH_c$ Co Based Films, (3) High ${}_iH_c$ Fe Based Films, (4) Films Which Exhibit a Large Degree of In the Film Plane Anisotropy, and (5) Films Which Exhibit Large Perpendicular Anisotropy.

(1) HIGH ENERGY PRODUCT Co BASED FILMS[1]

Different target configurations have been used to directly synthesize Sm-Co based films by selectively thermalized RF sputtering onto heated substrates in an in-plane magnetic field. Samples have been sputtered so that in certain cases a systematic gradient in Sm to Co concentration was created along the length of the substrates and in other cases so that films of a fixed uniform composition were deposited. Small subregions of a single substrate then serve as samples of a specific composition. For the Sm to Co composition range from 70 to 95 at. % Co gradient synthesized samples exhibit high magnetization and moderate intrinsic coercive forces. The maximum energy product of 20 MG-Oe was obtained for a sample with 90.6 at. % Co containing mostly the Sm_2Co_{17} phase. In addition samples were synthesized from uniform composition targets at certain fixed compositions. Films sputtered from uniform composition $Sm_2(Co,Fe,Zr)_{17}$ targets exhibited square in-plane hysteresis loops and static energy products up to 21.2 MG-Oe. A hysteresis loop for an as sputtered uniform composition $Sm_2(Co,Fe,Zr)_{17}$ film measured in the film plane and parallel to the field applied in the film plane, H-(s), during sputter deposition if shown in Fig. 1. It should be noted that this sample has received no subsequent heat treatment to the sputter deposition.

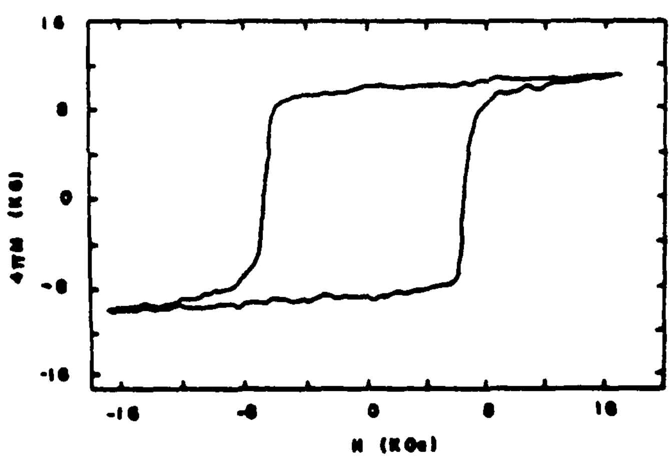


Fig. 1. Hysteresis loop measured in film plane parallel to H_s for a $Sm_2(Co,Fe,Zr)_{17}$ sample directly crystallized onto a heated substrate with field H_s applied during the sputter deposition.

It was observed that uniform composition $SmCo_5$ films could be grown which exhibited either the (200) or the (110) texture as a function of the sputtering rate. Relatively higher sputtering rates of approximately 5 Å/sec resulted in the (200) texture growth pattern as shown in Fig. 2 while lower rates resulted in (110) textured films as shown in Fig. 3. These differently textured $SmCo_5$ films differ in both their microstructure and magnetic properties. The (110) textured films are finer in microstructure and magnetically harder than the (200) films. The uniform composition $SmCo_5$ films of either texture in general were of smaller grain size and higher coercive forces than the $SmCo_5$ subregions from films sputtered with a composition gradient spanning the 1-5 phase.

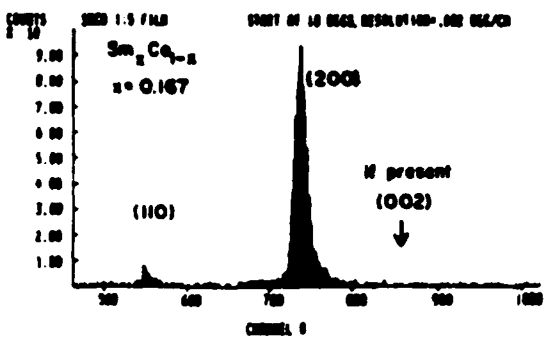


Fig. 2. X-Ray diffraction trace for the sample of Fig. 5, Si(Li) detector. The (100) and (300) reflections are present off the ends of this trace.

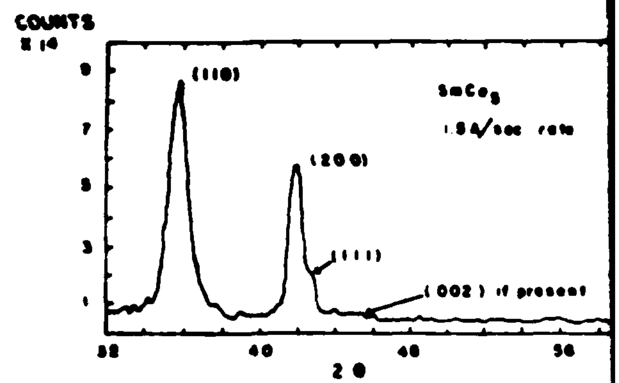


Fig. 3. X-ray pattern of a $SmCo_5$ sample sputtered using deposition 1.5 Å/sec from $SmCo_5$ targets onto heated substrate in the presence kOe filled in film plane.

A uniform composition $SmCo_5$ film sputtered at 1.5 A/sec which was (110) textured exhibited an intrinsic coercive force of 23 kOe and an energy product of 18 MG-Oe at -63 C. An in the film plane hysteresis loop measured parallel to H_s for magnetizing fields up to 88 kOe is shown in Fig. 4. This loop was measured at Fort Monmouth for us courtesy of Dr. Arthur Tauber and Dr. E. Potenziani. The high energy products reported here are due to both the high induction and to the squareness of the in-plane hysteresis loops when measured in the direction parallel to the field applied during the sputter synthesis.

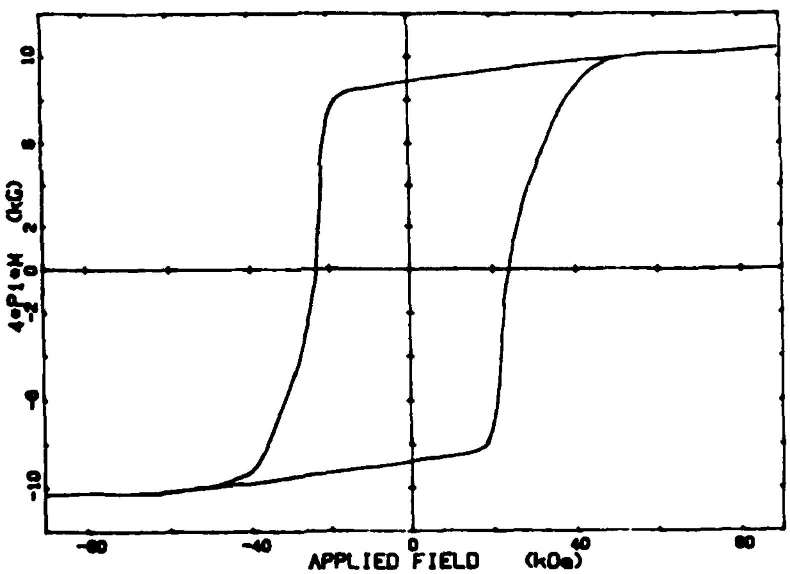


Fig. 4. In the film plane hysteresis loop measured parallel to H_s for a $SmCo_5$ sample directly synthesized at 1.5 A/sec.

(2) HIGH $_iH_c$ Co BASED FILMS[2,3]

Films which exhibit high $_iH_c$ values have been synthesized of the $SmCo_5$ compound. Films made from uniform composition targets have been directly crystallized onto heated substrates in the presence of an applied magnetic field, H_s, in the plane of the substrates. An in the film plane hysteresis loop for a sample of this type was shown in Fig. 4. Films of $SmCo_5$ synthesized by selectively thermalized sputtering at comparatively fast rates exhibit almost exclusively a (200) texture and exhibit in the film plane $_iH_c$ values of about 13 kOe. An in the film plane hysteresis loop for an as sputtered $SmCo_5$ film exhibiting (200) texture is shown in Fig. 5. Films synthesized by selectively thermalized sputtering at slower rates have been directly crystallized which exhibit a predominantly (110) texture. In this case in the film plane $_iH_c$ values as measured at room temperature are about 18 kOe.

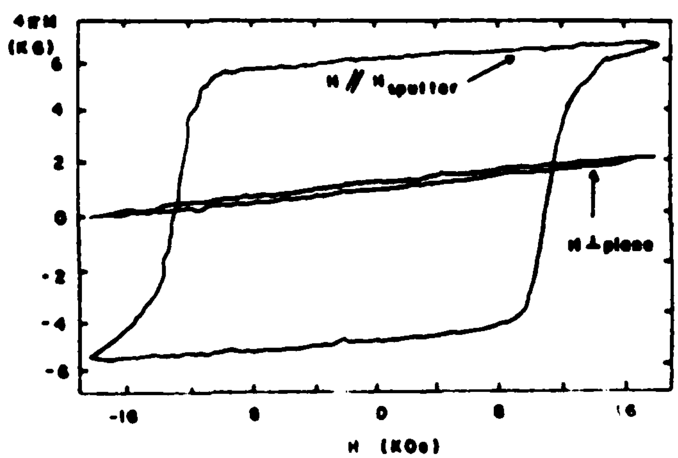


Fig. 5. Hysteresis loops both in film plane parallel to the 1.75 kOe field present during synthesis and perpendicular to the film plane for the $SmCo_5$ sample sputtered from $SmCo_5$ targets using a 5-Å/sec deposition rate onto a heated substrate.

The differences in texturing and in ${}_iH_c$ values for these films have been correlated with the different stacking sequences of the atoms for these textures. Films of $SmCo_5$ have also been synthesized by first depositing the material in an amorphous state and subsequently crystallizing the films insitu to achieve the $SmCo_5$ structure. Films of this type are premagnetized when they are removed from the system. Films of this type have been made which show littly variation in the in-plane moment for applied fields from -23 to +23 kOe. A minor hysteresis loop for applied fields up to 23 kOe for a film of this type is shown in Fig. 6. At present the maximum field that we use for measuring hysteresis loops at Queens College only goes to 23 kOe. X-Ray diffraction studies on crystalline $SmCo_5$ films that have been synthesized by this two stage process show that a reasonable degree of preferential alignment of the crystallite c-axes within the plane of the films has been achieved. The preferential alignment of the crystallographic axes in the film plane correlates with the appearance of a hard and easy axis of magnetization within the film plane. It is also observed, in contrast to the case of $SmCo_5$ films directly crystallized onto heated substrates, as shown in Fig.'s 2 and 3, that there is a certain concentration of crystallites with c-axis components normal to the film plane. This is believed to account for the relatively low 4*Pi*M values, 6 kG, observed for these films at 23 kOe applied in the film plane field. Note that the observed moment value is the value present when the films are removed from the sputtering system and that the application of fields to ± 23 kOe has been unable to change the observed moment. Higher values of applied fields are expected to further increase the available saturation moment nearer to the expected maximum for this system of 10 kG. X-Ray results on these and on amorphous films of Sm-Ti-Fe and $Nd_2Fe_{14}B$, which were grown with a magnetic field applies in the substrate plane, and which were subsequently crystallized in this same magnetic field, are due to be published shortly.[4]

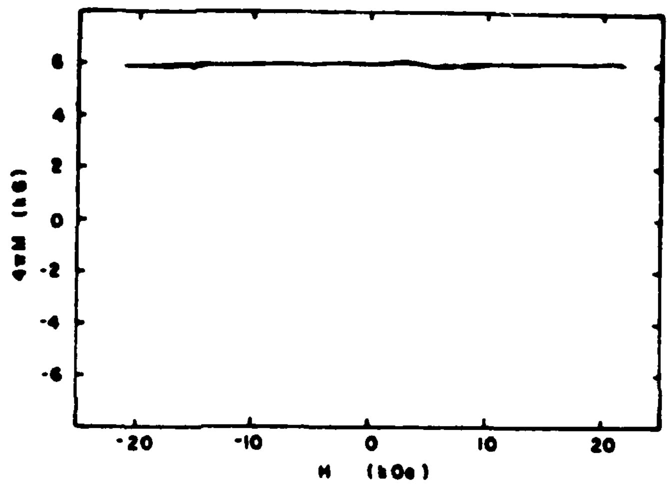


Fig. 6. In the film plane moment measured parallel to H_s for a $SmCo_5$ sample first made amorphous and then crystallized. The sample was premagnetized as removed from the sputtering chamber.

We hope to obtain high field data for such films shortly. This two stage synthesis procedure aids in achieving a fine grain structure which correlates with the increase in the ${}_iH_c$ values observed.

(3) HIGH $_iH_c$ Fe BASED FILMS[2,4,5,6]

Two different Fe based systems have been investigated which have been synthesized so as to exhibit high $_iH_c$ values at room temperature for in the film plane demagnetizing fields. One of these systems is the $Nd_2Fe_{14}B$ compound and the other is a new Sm-Ti-Fe system for which we recently reported in the film plane $_iH_c$ values at room temperature of 24 kOe.

Films of the $Nd_2Fe_{14}B$ compound have been directly synthesized by selectively thermalized sputtering and their magnetic properties have been measured at room temperature in applied fields up to 18 kOe. Films made at comparatively low substrate temperatures of 600 C show a larger moment perpendicular to the film plane than in the film plane for this range of magnetizing fields. Such a loop and the corresponding x-ray pattern are shown in Fig.'s 7 and 8 respectively. Films made at a higher temperature show a larger magnetization in the film plane than perpendicular to the plane for this range of magnetizing fields. The corresponding hysteresis loop and diffraction pattern is shown in Fig.'s 9 and 10. The change to easy axis in the film plane behavior correlates with the changes in the film texturing which shows that at the higher substrate temperatures the c-axis is more aligned into the film plane. Films made at higher substrate temperatures show only minor hysteresis loops when measured to fields of 18 kOe. A minor hysteresis loop of this type is shown in Fig. 11. The in the film plane intrinsic coercive force,

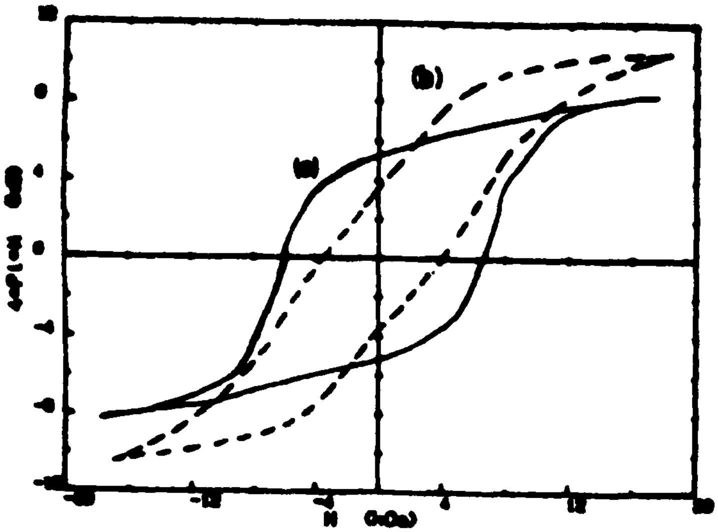


Fig. 7. Hysteresis loops measured in the film plane (a), and perpendicular to the film plane (b), for $Nd_2Fe_{14}B$ directly crystallized at 600 C.

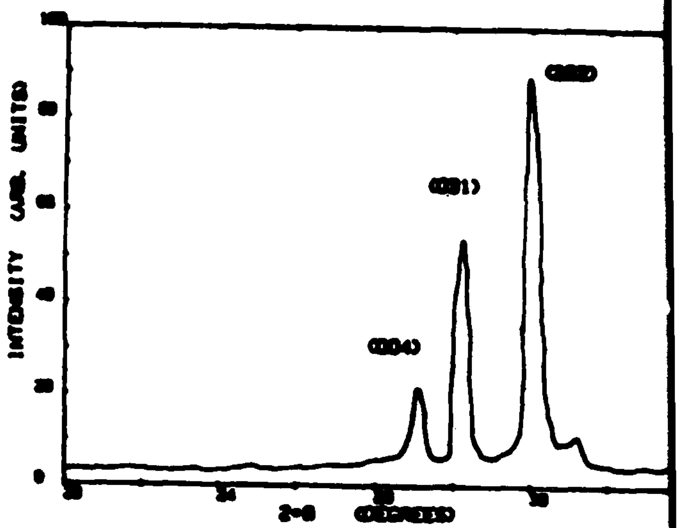


Fig. 8. X-Ray diffractometer trace, Cu radiation, of $Nd_2Fe_{14}B$ film made at 600 C.

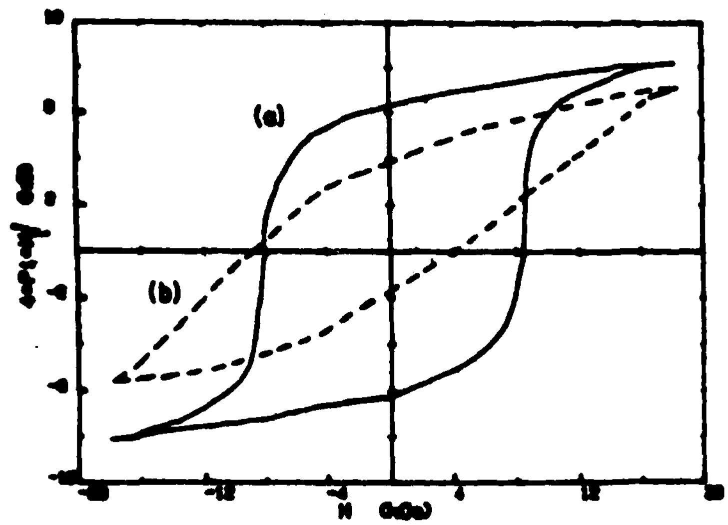


Fig. 9. Hysteresis loops measured in the film plane (a), and perpendicular to the film plane (b), for $Nd_2Fe_{14}B$ directly crystallized at 700 C.

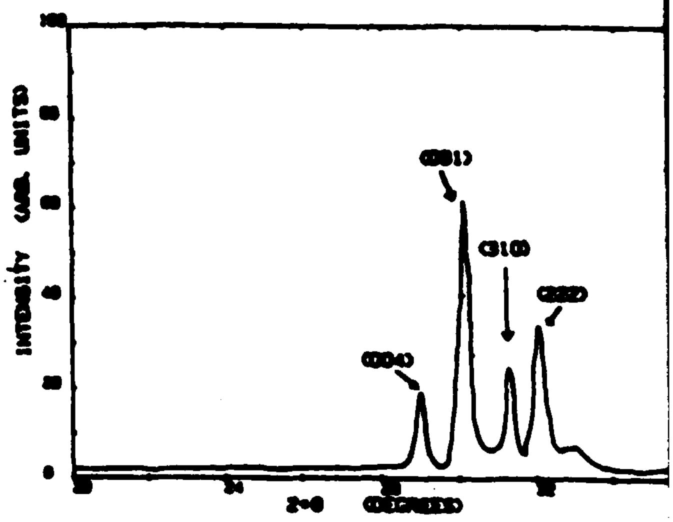


Fig. 10. X-Ray diffractometer trace, Cu radiation, of $Nd_2Fe_{14}B$ film made at 700 C.

$_iH_c$, is at least 16 kOe as deduced from such minor loops. We have as yet not been able to get high field data for such samples.

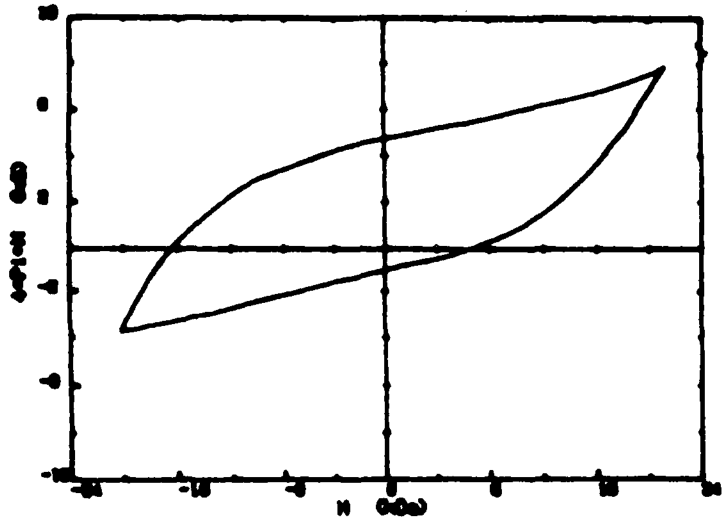


Fig. 11. Minor hysteresis loop measured in the film plane for $Nd_2Fe_{14}B$ directly crystallized at 750 C.

Recently films of $Nd_2Fe_{14}B$ which exhibit large values of the effective perpendicular anisotropy constant have been synthesized. The magnetization when measured perpendicular to the film plane does not drop until appreciably large reverse fields are applied. More will be said about these films in the section covering perpendicular anisotropy films in Section 5.

A phase which contains only Sm, Ti, and Fe has been synthesized in film form which exhibits an in the film plane intrinsic coercive force, $_iH_c$, of 24 kOe at room temperature. In August 1985 we were able to get some high field measurements on such a sample measured at the National

Magnet Laboratory courtesy of the Grumman Corporation. The central part of such a hysteresis loop is shown in Fig. 12. This hysteresis loop was measured on a sample which was first made amorphous with a magnetic field applied in the substrate plane during deposition and then crystallized in the presence of this same field. The magnetic properties and the crystal structure did not correspond precisely to the films which had been made directly crystalline onto heated substrates. The x-ray pattern for the high ${}_iH_c$ films which had been crystallized from amorphous films corresponded to a $Sm_2Fe_{14}Ti$, a = 8.285 A and c = 12.500 A, analogue of the $Nd_2Fe_{14}B$ phase. We are now attempting to make single phase samples of this type. We believe we now know how to do this. High field measurements will be required to keep track of changes in the magnetic properties produced by changes in the sputtering parameters. Progress in this direction would be greatly aided by having high field measurement capability available at Queens College.

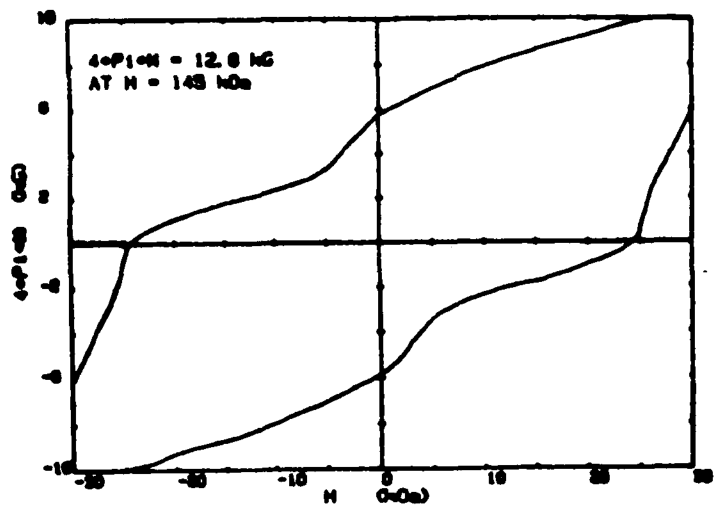


Fig. 12. Central region of room temperature high field hysteresis loop measured in the film plane and parallel to H_s for a $(Sm+Ti)Fe_5$ sample. This sample contained no Co.

Many samples of this basic Sm-Ti-Fe system have been made by slightly different methods which show moments up to 16 kG when magnetized to 18 kOe perpendicular to the film plane, but only show moment values up to about 10 kG for the same field applied in the film plane. High field data will also be necessary to characterize the magnetic properties of these samples. A sample of this type is shown in Fig. 13. These samples were made by selectively thermalized sputtering as a part of a study of $(Sm+Ti)(Fe,Co)_5$ samples in which the relative amounts of Sm to Ti as well as Fe to Co ratios have been varied. The use of Ti has allowed the synthesis of $(Sm+Ti)(Fe,Co)_5$ films with up to 33 at.% Fe replacement for Co. Films of this type have been synthesized which exhibit an in the film plane energy product of 15 MG-Oe.

Recently a patent was issued to the U. S. Army Research Office under Patent No. 4547276, date of issue: October 15, 1985, for the synthesis of a $(Sm+Ti)Fe_5$ compound. The title of this patent is "Method of Directly Crystallizing a (Sm + Ti):Fe = 1:5 Compound". The inventor is Fred J. Cadieu.

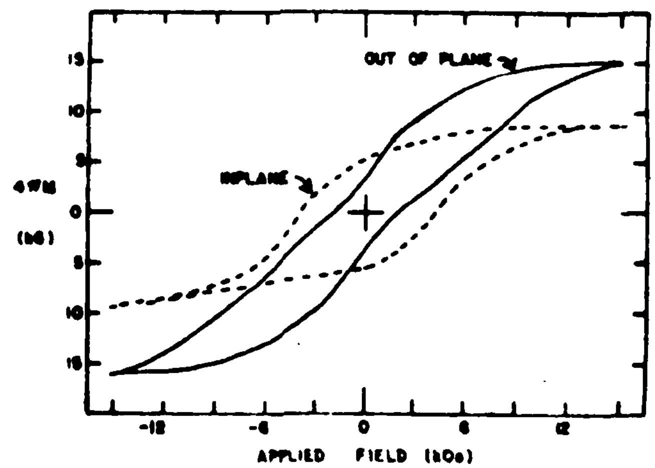


Fig. 13. Hysteresis loops at room temperature for a Sm,Ti,Fe(0.095:0.046:0.860) sample synthesized onto 600 C substrate. Loops for the magnetometer field perpendicular to film plane and in the film plane parallel to H_s are shown.

(4) FILMS WHICH EXHIBIT A LARGE DEGREE OF IN THE FILM PLANE ANISOTROPY[7]

For amorphous compositions corresponding to the $SmCo_5$ compound, and for the $Sm_2(Co,Fe,Zr)_{17}$ composition, as well as for amorphous compositions of $(Sm+Ti)Fe_5$, samples have been made which exhibit a large degree of in the film plane anisotropy. This anisotropy refers to an easy and hard axis in the film plane at right angles to each other. The easy axis is in the direction of the H_s field. Anisotropy constants up to $2.7 \times 10^{+6}$ erg/cm^3 have been measured. Such a set of hysteresis loops for a $(Sm+Ti)Fe_5$ composition with a Ti:Fe atomic composition ratio of 1:9 is shown in Fig. 14. It should be noted that the remanent moment value is nearly equal to the saturation moment. The values of 4*Pi*M, and the in the film plane anisotropy constants observed for several different amorphous compositions are shown in Table I. Such films are possible candidates for digital longitudinal recording.

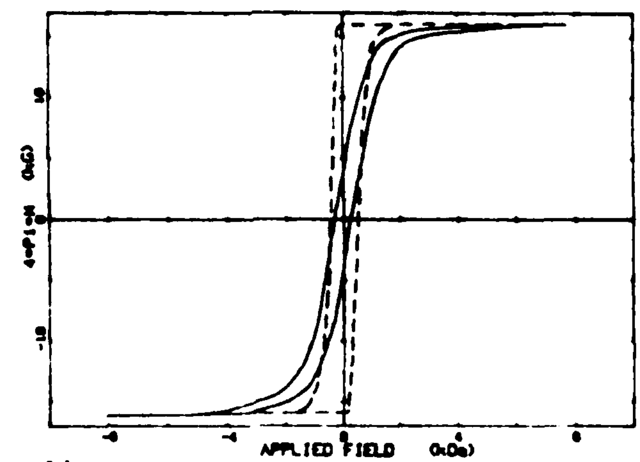


Fig. 14.

In-plane hysteresis loops parallel, rectangular, and perpendicular to *H* sputter, 2.5 kOe, for a SmTiFe (0.075: 0.092: 0.833) amorphous composition.

TABLE I. The saturation moments, $4\pi M_s$, in-plane anisotropy fields, H_A, and in-plane anisotropy constants, k_A, for the different amorphous compositions in the as sputtered state. For the first three compositions, H_s = 2.5 kOe, and for the last, H_s = 1.75 kOe.

	$4\pi M_s$ (± 5%) (kG)	H_A (± 5%) (kOe)	k_A (± 10%) (10^{+6} erg/cm^3)
$SmCo_5$	10.3	10.6	2.6
$Sm_2(CoFeZr)_{17}$	15.2	5.8	2.2
SmTiFe (0.075, 0.092, 0.833)	16.4	5.0	1.5
SmTiFe (0.069, 0.098, 0.833)	14.2	3.1	1.4

(5) FILMS WHICH EXHIBIT A LARGE PERPENDICULAR ANISOTROPY[5,6,8]

Film samples of a new $Sm(Fe,Ti)_2$ system have been synthesized which exhibit a large perpendicular anisotropy of $4x10^6$ erg/cc. Our fabrication procedure has involved two steps: (1) sputtering the films in an amorphous state by selectively thermalized sputtering, and then (2) crystallizing the films to achieve a new perpendicular anisotropy system. A set of hysteresis loops for samples of this new compound are shown in Fig. 15. We have also recently observed large values for the effective perpendicular anisotropy constant for specially prepared samples of the $Nd_2Fe_{14}B$ compound. In this case values of K effective perpendicular up to 8 x 10^{+6} erg/cm^3 have been observed at room temperature.

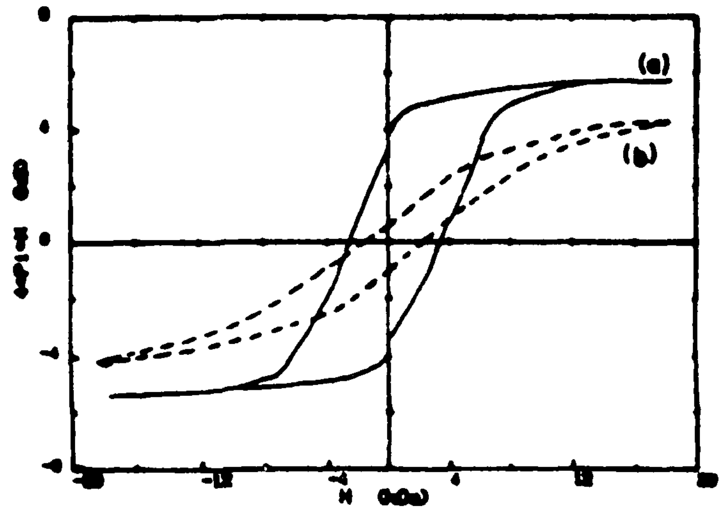


Fig. 15. Hysteresis loops measured perpendicular (a), and parallel to film plane (b), for a $Sm(Ti,Fe)_2$ sample.

Films of the $Nd_2Fe_{14}B$ compound have recently been synthesized which exhibit large values of the effective perpendicular anisotropy constant. A fuller report on these results was made at the Intermag 86 Conference which was held in Phoenix, April 1986. A minor hysteresis loop as measured perpendicular to the film plane is shown in Fig. 16 without and with demagnetization corrections. This data is unusual in that the magnetization even when measured perpendicular to the film plane does not drop appreciably until well into the 2nd quadrant. High field data will be needed to fully saturate this sample which may explain why the magnetization at 20 kOe applied field is less that the expected saturation value. Hopefully high field data can be measured at Fort Monmouth in February 1986. An x-ray diffractometer trace with Cu radiation for this sample is shown in Fig. 17.

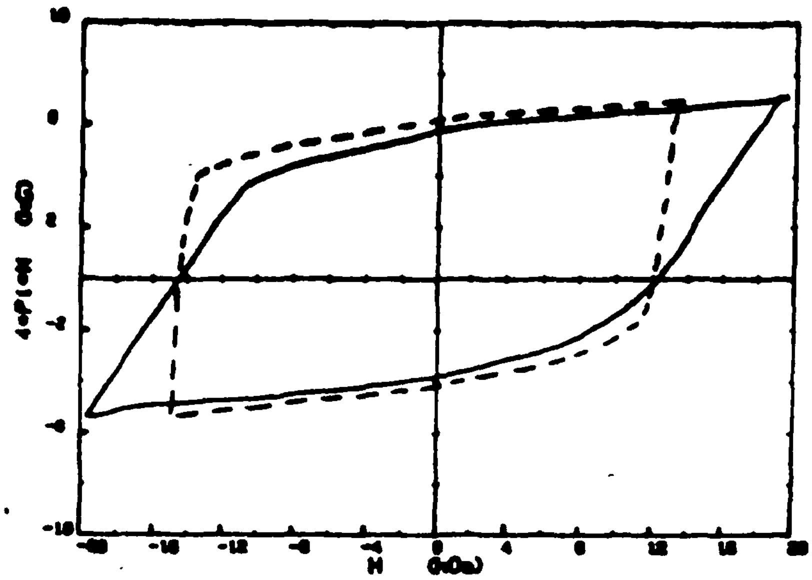


Fig. 16. Solid line perpendicular 4PiM versus applied H. Dashed line 4PiM versus internal effective field, after correction for N_d.

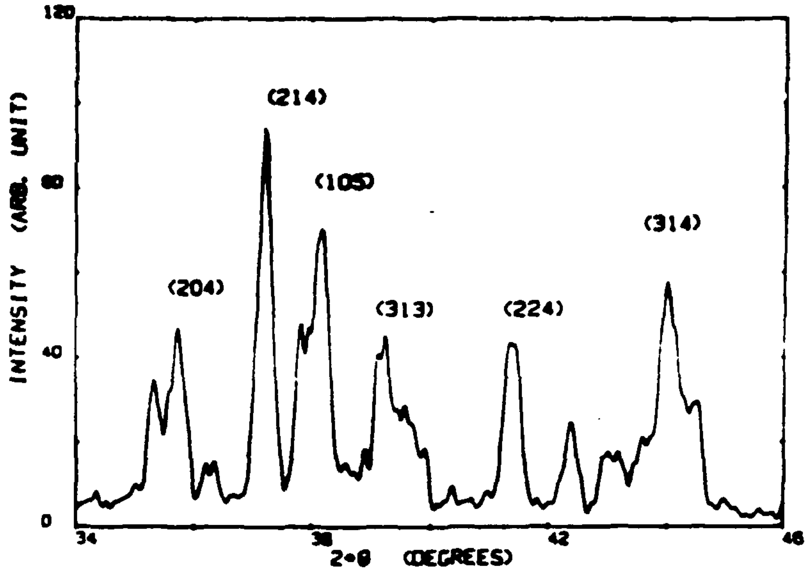


Fig. 17. X-Ray diffraction pattern for the sample of Fig.16, Cu radiation.

The lattice parameters based on tetragonal indexing are determined by minimizing a function

$$F = (1/\sin(22.5)*\tan(22.5))*<(1/N)\mathrm{SUM}[(A_{exp} - A_{calc})*\sin(A)*\tan(A)]^2>^{1/2},$$

where the sum is over N observed reflections at the angles 2A. The function F forms a surface of decreasing elliptical cross sections as a function of the a, and c, lattice parameters. Cuts through the minimum value of F for constant a and c are shown in Fig.'s 18 and 19. The experimentally determined best values of a and c lattice parameters are: a = 8.770 ±0.003 A and c = 12.278 ±0.005 A. The calculated x-ray density based on these lattice parameters is 7.604 ±0.013 gm/cm^3.

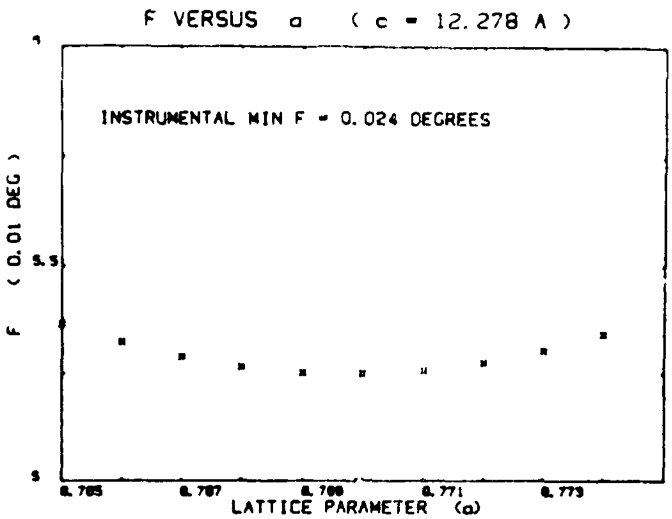


Fig. 18. Section through F surface at minimum point for constant c = 12.278 A.

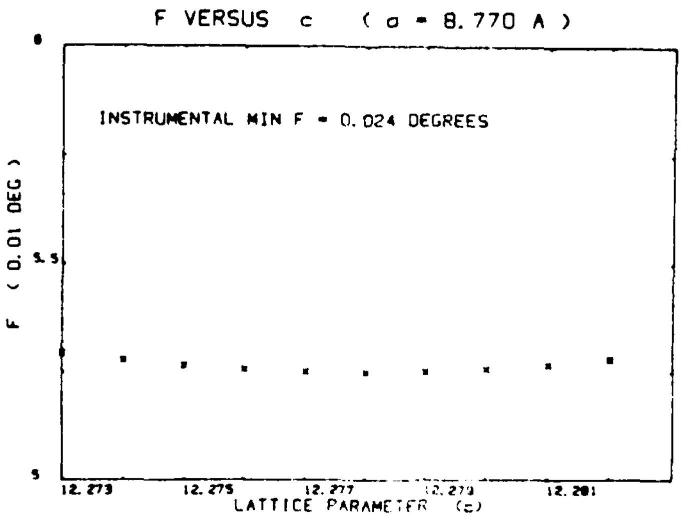
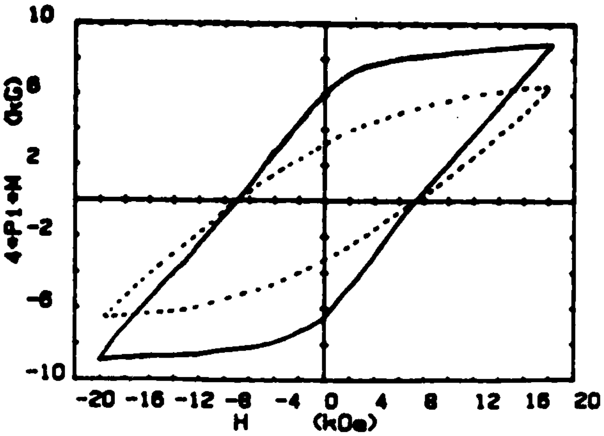


Fig. 19. Section through F surface at minimum point for constant a = 8.770 A.

Among the rare earth transition metal permanent magnet systems, the $Nd_2Fe_{14}B$ bulk system exhibits the highest energy product of 45 MGOe. High energy product Co based materials that we have sputtered usually align the easy axis onto the substrate planes. Good examples are the $SmCo_5$ sputtered films which we have studied extensively [1]. Perpendicular anisotropy requires the easy axis to be normal to the film plane and is generally not possible if the saturation magnetization is very large. For the moment to align perpendicular to the film plane it has to overcome the demagnetization energy $2*Pi*M^2$. Perpendicular anisotropy crystalline phases, for example: Fe_xN, $Sm(Ti,Fe)_2$, and Co-Cr, exhibit either low or moderate saturation fluxes and lose the perpendicular anisotropy as the flux increases.[2,3,4] It is therefore surprising that $Nd_2Fe_{14}B$ film magnets can be sputtered with large effective perpendicular anisotropy constants of 1.2×10^7 erg/cc by controlling the sputtering parameters in spite of its large saturation flux. In the analogous Sm-Co system, only the amorphous $SmCo_3$ phase can be sputtered with perpendicular anisotropy. We had sputtered from 2 Co and 1 Sm elemental targets and directly synthesized the $SmCo_3$ crystalline phase onto heated substrates. Perpendicular remanent moment of 5 kG and coercivity of 6 kOe was observed. Details of these results will be discussed briefly [5]. Sputtering from uniform $SmCo_3$ targets reported earlier gave a perpendicular remanent moment of 2.9 kG and coercivity of 8 kOe [6]. The high energy product $SmCo_5$ hexagonal phase cannot be sputtered with perpendicular anisotropy. In contrast, the $Nd_2Fe_{14}B$ complex tetragonal phase gives more degrees of freedom in crystal orientation that can be controlled by varying the sputtering condition. It has been found that increased substrate deposition temperatures up to 850 C increase $_iH_c$. It has been observed that for deposition rates less than 1.8 A/sec the films exhibited the easy axis of magnetization perpendicular to the film plane, while for deposition rates greater than 1.8 A/sec the easy magnetization axes was in the film plane. This is a modified correlation to that based on less data we had reported earlier.[7]

The hysteresis loop measured perpendicular to the film plane for an as sputtered $Nd_2Fe_{14}B$ film deposited at 700 C substrate temperature with a sputtering rate of 0.9 A/sec is displayed in Fig. 20 (solid line). The observed moment at 18 kOe was 9.2 kG. This combined with a relative low coercivity of 7.3 kOe gave an inclined shape to the otherwise square loop because of the demagnetization factor of 4*Pi. The dash line indicates measurements parallel to the film plane. The remanent moment of the dashed line is 3.5 kG compared to 6.2 kG for the solid line indicating that the easy axis of magnetization is more aligned perpendicular to the film plane. The x-ray diffraction is shown in Fig. 21 where it is observed that the (105) and (214) are the prominent reflections suggesting a preferential texturing of the c-axis which correlates with the easy magnetic axis being normal to the film plane.



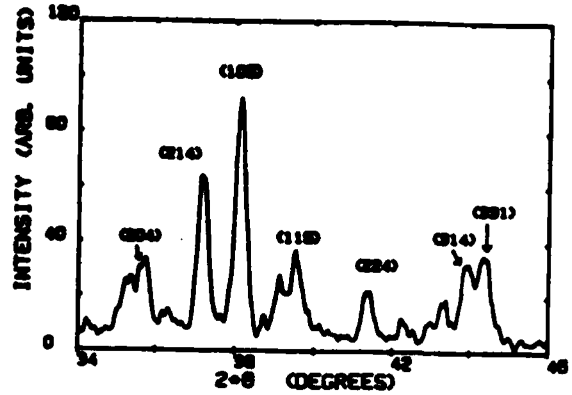
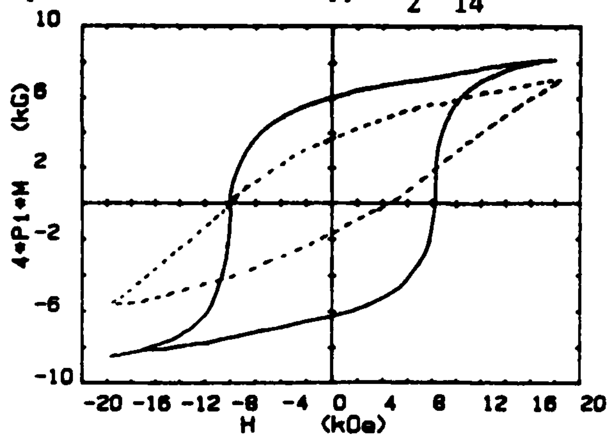


Fig. 20. Hysteresis loops measured normal to the film plane (solid line) and in the film plane (dotted line) for Nd-Fe-B directly crystallized at 700 C with 0.9 A/sec sputtering rate.

Fig. 21. X-Ray diffraction trace, Cu radiation, for sample of Fig. 20.

Films sputtered under the same conditions, but at a higher rate, resulted in films with the c-axis preferentially aligned onto the film plane. In Fig. 22 the magnetic responses of a $Nd_2Fe_{14}B$ sputtered under the same condition as the film in Fig. 20 but with a rate of 2 A/sec is shown. The parallel to the film plane direction attained a higher moment compared to the normal to plane direction. This suggests that the c-axis is more aligned onto the film plane. This is consistent with the x-ray diffraction pattern as shown in Fig. 23 where the (031) line is larger than the (222) line. The increased intensity of the (310) line also indicates that the c-axis is aligned more onto the film plane.[7] The most striking difference of Fig.21 and Fig.23 is that most major peaks occured at angles from 34 to 46 degrees in Fig.21 whereas most major peaks occured at lower angles from 28 to 33 degrees in Fig. 23. These correspond to d-spacings of 1.973 A to 2.637 A in the low rate sputtered film and 2.715 A to 3.187 A in the high rate sputtered film. This illustrated that low rate sputtering produced a relatively closer packed stacking sequence than high rate sputtering. This is again consistent with the fact that low rate sputtering produces a c-axis preferential texture normal to the film plane since the c-axis is the more closely stacked direction in the $Nd_2Fe_{14}B$ tetragonal phase. The low rate sputtering allows relatively more Ar atom bombardment during the growth process when compared to the sputtered atoms flux. This Ar pseudo compression favors the closer packed stacking sequence. In high rate sputtering where the c-axis was more aligned onto the film plane, this was also consistent with the lattice exhibiting predominantly larger d-spacings so as to minimize the stored magnetic energy. Therefore it is concluded that low rate sputtering is essential in producing perpendicular anisotropy $Nd_2Fe_{14}B$ films.



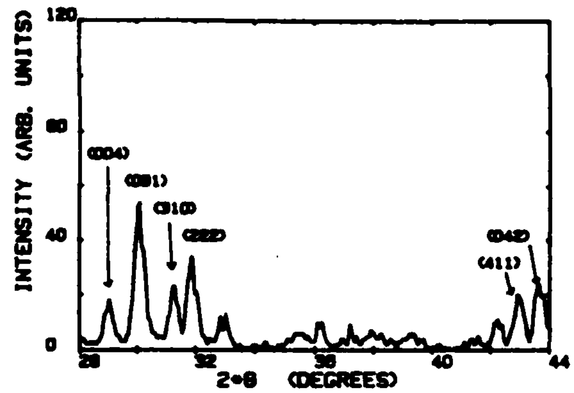
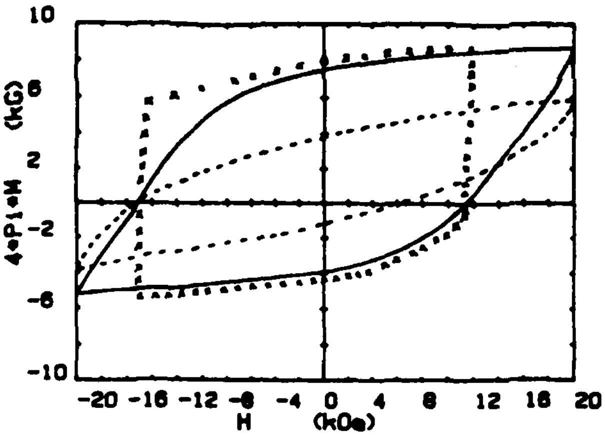


Fig. 22. Hysteresis loop measured in the film plane (solid line) and perpendicular to the film plane (dotted line) for Nd-Fe-B directly crystallized at 700 C with 2 A/sec sputtering rate.

Fig. 23. X-Ray diffraction trace, Cu radiation, for the sample of Fig. 22.

The hysteresis loop measured perpendicular to the film plane for an as sputtered $Nd_2Fe_{14}B$ film deposited at 800 C substrate temperature with a 0.6 A/sec sputtering rate is displayed in Fig. 24 (solid line). The flux at 20 kOe external field was 8.5 kG and the perpendicular remanent at zero applied field was 7.5 kG. The ${}_iH_c$ was 14.8 kOe as a result of the higher deposition temperature. The magnetization versus internal effective field, after correction for the demagnetization factor, is shown as cross points. The high ${}_iH_c$ and remanent moment makes these film magnets unique among the perpendicular anisotropy materials. The area bounded by the magnetization curves in the easy (solid line) and hard (dash line) directions gave an effective perpendicular anisotropy constant of 1.2 x 10^7 erg/cc. An x-ray diffraction trace is shown in Fig.25. The prominent peaks were the (214), (204), and (105) reflections. This indicated a preferential texturing of the c-axis normal to the film plane. When compared to the sample of Fig. 21, this sample also showed more c-axis texturing normal to the film plane.



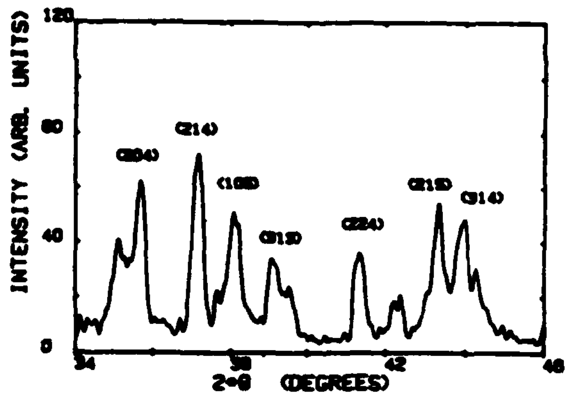


Fig. 24. Hysteresis loop measured normal to the film plane (solid line) for Nd-Fe-B directly crystallized at 800 C with 0.6 A/sec sputtering rate. The cross points show 4*Pi*M versus internal effective field after correcting the solid curve for the demagnetization factor (normal to film plane).

Fig. 25. X-Ray diffraction trace, Cu radiation, for the sample of Fig. 24.

One of the conclusions reached from these studies was that low rate sputtering is essential to produce perpendicular anisotropy in $Nd_2Fe_{14}B$ films, while rates greater than 1.8 A/sec resulted in films with in the plane anisotropy. High deposition temperatures have been required to synthesize high coercivity in this system. The substrate temperature versus ${}_iH_c$ is shown in Fig. 26 for deposition rates from 0.44 A/sec to 3 A/sec. The ${}_iH_c$ values increase with increasing deposition temperatures for perpendicular and in the plane anisotropy films.

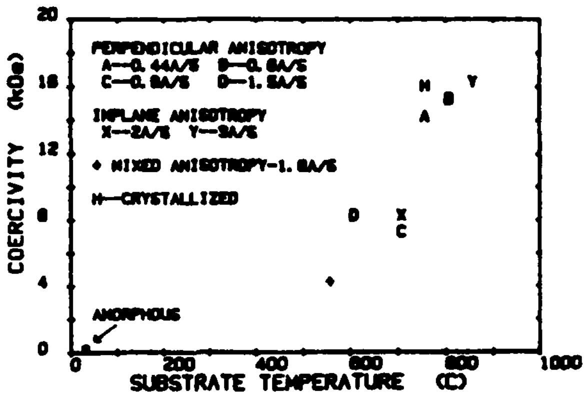


Fig. 26. The coercive force versus substrate deposition temperature for $Nd_2Fe_{14}B$ films synthesized at different rates. Also shown are the ${}_iH_c$ values for an amorphous film before and after crystallization.

For coercivities greater than 13 kOe only minor loops have been observed for the maximum magnetizing field of 20 kOe. The as measured remanent 4*Pi*M perpendicular to the plane for a Nd-Fe-B film made at 0.44 A/sec with a ${}_iH_c$ value of 14 kOe is reported as 9.3 kG. For higher initial magnetizing fields this value is expected to increase. The $Nd_2Fe_{14}B$ system is unique in that large values of the static energy product and extreme values of effective perpendicular anisotropy constants can be obtained in the same compound. The sputtering rate has been used as an indicator of preferential c-axis texturing out of the film plane which increases with decreasing rates. The precise dependence of c-axis texturing versus sputtering rate is expected to be sputtering system dependent. The ratio of the intrinsic perpendicular anisotropy constant (15×10^6 erg/cc) to the demagnetization energy ($2*Pi*M^2 = 2.8 \times 10^6$ erg/cc) gives a Q value of 5.3 .

The relatively lower ${}_iH_c$ of $SmCo_3$ gave a sharp drop to the magnetization in the second quadrant, Fig. 27 as compared to the Nd-Fe-B film of Fig. 25. It is concluded that Nd-Fe-B is the best known candidate for producing high ${}_iH_c$ perpendicular anisotropy films.

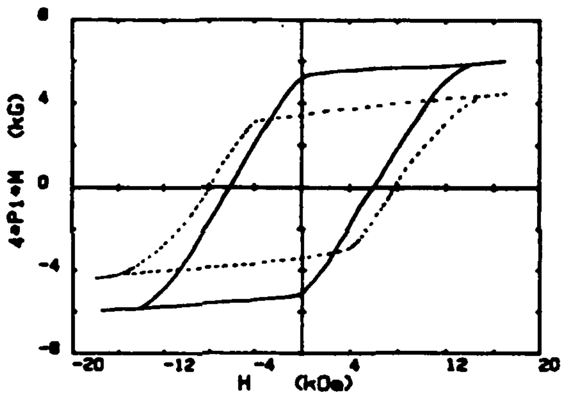


Fig. 27. Hysteresis loops measured perpendicular to the film plane, solid curve, and in the film plane, dotted curve, versus applied field, H, for a crystalline $SmCo_3$ film directly crystallized at 650 C substrate temperature.

MICRON SCALE DEVICE GEOMETRIES[9,10]

A method has been devised which allows the construction of miniature periodic permanent magnet arrays with a repeat length in the 1 to 200 micron range (1 micron = 10^{-6} meter). Such arrays can be constructed by utilizing either alternating layers or regions of permanent magnet film materials with different coercive forces. For example alternating layers of $SmCo_5$ can be deposited such that a magnetic layer of $SmCo_5$ with a (200) crystal texture and then (110) layers are deposited. We have shown that the (110) textured films can be synthesized with an intrinsic coercive force, ${}_iH_c$, of approximately 15 to 23 kOe at room temperature. The $SmCo_5$ intervening layers would have a lower ${}_iH_c$ value as we have shown. It may or may not be useful in certain cases to use spacer layers of nonmagnetic film layers between the $SmCo_5$ (200) and (110) layers. When such a layered structure is exposed to a strong in the film plane magnetic field all layers will be saturated by a sufficiently strong field of approximately 40 to 60 kOe. When a reverse in the film plane field of approximately 12 kOe is then applied to this layered device only the (200) layers will have their magnetic moments reversed. After the removal of all external magnetic fields, the layered structure will have the (200) and (110) textured layers magnetized in opposite directions. If the $SmCo_5$ (200) and (110) each have a thickness of 10 microns, the magnetic period of such a device will be 20 microns plus the thickness of any nonmagnetic layers such as intervening spacer layers. The novel feature of this invention is the use of magnetic materials with different intrinsic coercive forces so that adjacent layers can become magnetized in opposite directions through the use of external fields whose spatial extent is large compared to the size of the regions being magnetized. This is to be contrasted to writing on a magnetic tape material where the magnetized head to reverse a small region of tape has to also have a physically small write head size.

These principal materials are of interest: (1.) Square hysteresis loop films of the compound $SmCo_5$ exhibiting a (200) predominant texture and intrinsic coercive forces, ${}_iH_c$, of approximately 8 kOe, (2.) Square hysteresis loop films of the compound $SmCo_5$ exhibiting a (110) predominant texture and ${}_iH_c$ values of approximately 15 kOe, and (3.) Square hysteresis loop films of the compound $Sm_2(Co,Fe,Zr)_{17}$ of the TDK type exhibiting ${}_iH_c$ values of approximately 7 kOe. Film layers of the $Nd_2Fe_{14}B$ compound synthesized so as to exhibit either in the film plane anisotropy or perpendicular to the film plane anisotropy. Film layers of $Sm(Ti,Fe)_2$ which exhibit perpendicular anisotropy.

The arrays can be constructed in several different manners. In method (1.) alternating layers of the compound $SmCo_5$ with the (110) texture and the (200) texture are deposited over each other. The thickness of the layers can be varied from the range of approximately 1 micron to several hundred microns. When this layered system is saturated in the film plane and preferably parallel to the direction in the film plane in which the field H_s was applied, all layers will be saturated by fields of approximately 23 or more kOe. The initial magnetizing field should now be reversed and raised to a level of approximately 12 kOe so that only the moments in the lower ${}_iH_c$ layers of $SmCo_5$ having the (200) texture will be reversed. All external fields are now removed. The resulting physical structure is now composed of alternating layers of $SmCo_5$ of the (200) and (110) textures magnetized such that the magnetization is oppositely directed in successive layers. The expected hysteresis loop of such a structure when measured in the plane of the film layers and parallel to the direction of initial magnetization is as shown in Fig. 28. In method (2.) alternating layers of the compound $SmCo_5$ with the (110) predominant texture and layers of $Sm_2(Co,Fe,Zr)_{17}$ are deposited over each other. When magnetized initially and then the lower coercive force layers reversed in magnetization as for the method (1.) structures, the resulting magnetization state will again be one of oppositely directed moments in successive layers. The saturation moments in the successive layers will, however, not be the same since the different compounds have slightly different saturation moments. It may be desirable in this case to make the layers of $SmCo_5$ slightly thicker than the alternating layers of $Sm_2(Co,Fe,Zr)_{17}$ to compensate for the lower saturation moment of the $SmCo_5$ compound. Although thin layers can be made so that the magnetic field from one layer interacts strongly with that of the next layer, the object here is to make each layer act independently, but to be physically adjacent to each other. Films the order of 1 micron in thickness would satisfy this criterion, but probably thicknesses rangeing up to several hundred microns would be more useful. We have had no problems with films of 20 to 30 microns peeling from substrates and see no problems in making thicker alternating layers. The remanent magnetic induction, B_r, observed in individual film layers is approximately 9-11 kG.

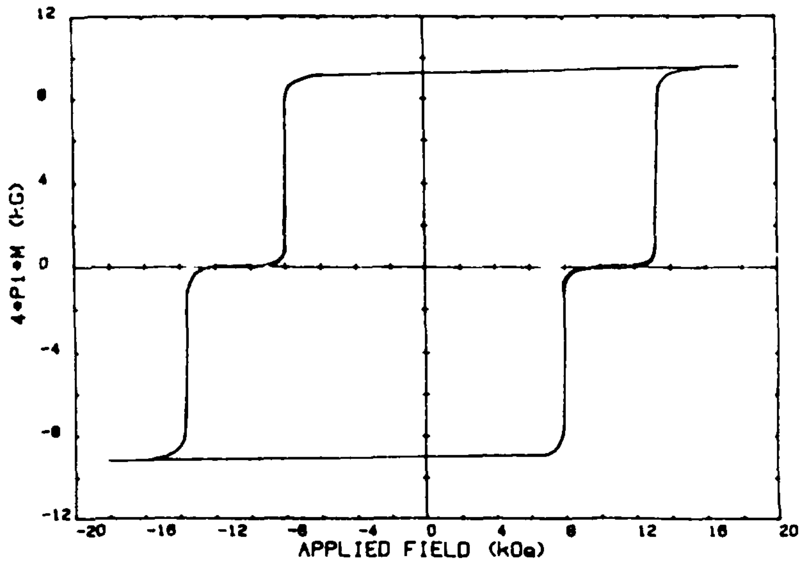


Fig. 28. An idealized hysteresis loop for a set of alternating film layers of $SmCo_5$ with (200) and (110) textures is shown for the magnetometer field applied in the plane of the film layers.

Such miniature periodic permanent magnet arrays become useful as an undulator structure when the periodically reversing magnetic field from successive layers is used to alternately deflect an electron beam. Periodic arrays constructed from bulk magnets have been used in the construction of free electron lasers and at most synchrotron sources for the generation of coherent radiation. To use these arrays as an undulator an electron beam is passed through a hole in the layers and substrate or through a patterned structure formed from the layers. The electron beam as it passes through the B field from successive layers is alternately deflected in a plane perpendicular to the path of the beam and the B field. The B field in the micron scale undulator structure as described is oppositely directed in directions perpendicular to the path of the electron beam.

The possibility of making small scale magnet geometries by film synthesizing film magnet layers has also been considered. These considerations are in a preliminary stage of development. A cladded thin film magnet geometry is possible in which a layer such as a high remanent moment $Nd_2Fe_{14}B$ layer which is first deposited onto a substrate and then a layer of Sm-Co based material is deposited over this. The Sm-Co based layer exhibits in the film plane anisotropy. A gap region is constructed by placing two similarly coated substrates next to each other so as to form a gap region.

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Method of Making a Miniature Scale Periodic Permanent Magnet Array and Miniature Scaled Periodic Permanent Magnet Array So Formed, Inventors: F. J. Cadieu and T. D. Cheung, Fort Monmouth CECOM Docket No. D-4170, patent application to be filed by U. S. Army Research Office and Fort Monmouth CECOM, September or October 1986.

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